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Abstract: We present photoluminescence measurements under strong magnetic field done on a sample with charged (n-doped) quantum dots. We show that the broadening of the luminescence line does not give a measure of the QDs size dispersion, and that the coupling of electron/hole with LO phonons pairs is greatly enhanced, because of the presence of the ionised impurities nearby the charged dots.

PACS: 73.21.La, 73.40.Kp, 78.67.Hc
Keywords: Quantum dots, optical properties, doping

1. Introduction

Various experimental and theoretical works demonstrate that electrons confined in quantum dots (QDs) are strongly coupled to the longitudinal optical (LO) vibrations of the underlying semiconductor lattice [1]. This coupling leads to the formation of the so-called quantum dot polarons, which are the true excitations of a charged dot. The infrared (FIR) absorption probes directly the polaron levels instead of the purely electronic ones. The intra-band absorption spectrum of an ensemble of charged dots clearly display thin (≈ few meV) features [1,6], despite the unavoidable statistical distribution of the dot size.

For electron-hole pairs confined in a QD, the coupling to optical phonons manifests itself in two ways: by an important modification of their energy levels, due to the formation of excitonic polarons, and by the appearance of phonon replica in their photoluminescence [2-5]. However, inter-band transitions of semiconductor QDs are inhomogeneously broadened because of fluctuations of the confining potentials. In particular, the usual broad-bell-like form of the non-resonant photoluminescence (NRPL) of a dot ensemble is generally taken as a measure of its size inhomogeneity. Selective (or resonant, in energy) excitation allows to partially circumvent this difficulty, since only a fraction of the available QDs may resonantly absorb incoming photons with energy below the WL edge. In particular, resonant photoluminescence (RPL) experiments done on weakly inhomogeneous samples (FWHM ≤ 25 meV) have shown the existence of low energy replica, and allowed an estimation of the Huang-Rhys parameter (which gives a measure of the strength of the coupling to optical phonons) [2,4]. We present in this work NRPL and RPL measurements under strong magnetic field done on a sample with charged (n-doped) quantum dots. We show that the broadening of the luminescence line does not give a measure of the QDs size dispersion, and that the coupling of electron/hole pairs with LO phonons is greatly enhanced, because of the presence of the ionised impurities nearby the charged dots.

2. Experimental results

We have done low temperature (T = 4K) interband RPL and NRPL measurements on an ensemble of n-doped InAs/GaAs dots (doping ≈ one electron per dot) under strong magnetic field (up to B = 28 T) applied along the growth axis. The QD electron filling was realised by a delta doping of the GaAs barrier at 2 nm under the dot layer. More details on the sample and growth characteristics are given in ref. [6]. Figure 1 shows the NRPL spectra obtained by exciting weakly in the GaAs barrier at B = 0 and 28T (the same spectra are obtained when decreasing the excitation intensity by one order of magnitude). The spectra are significantly broader (≈ 80 meV) than usually observed (≤ 35 meV) for undoped dots obtained by similar growth conditions. At first glance, such a large broadening appears incompatible with the narrow lines of FIR experiments. It turns out that the NRPL width is essentially governed by the height dispersion. It is nevertheless difficult to explain why the doping would affect the QDs characteristics, so that our n-doped dots would have substantially larger height dispersion, as compared to undoped dots of same average sizes and grown under similar conditions. In the following we show that this is actually not the case: the presence of the doping impurities nearby the dots strongly affects the NRPL profile, but its *intrinsic* inhomogeneous width is only slightly affected by the doping. In other words, and contrarily to undoped QDs, for n-doped samples the width of the NRPL spectrum does *not* give a correct measure of the DQs size inhomogeneity. In addition, we show in the following that this difference is due to two effects, and, in particular, because the coupling of bound carriers to optical phonons is enhanced in n-doped samples as compared to undoped ones. As discussed below, these conclusions result from the analysis of the RPL spectrum and of its variation in the presence of an applied magnetic field.

Figure 2 shows the magnetic field dependence of the RPL spectra by exciting below the wetting-layer edge at $\hbar\omega_{in}$ = 1292 meV. Let us consider initially the zero field case. We observe a weak luminescence for detunings up to ≈ 52 meV, while a series of roughly equidistant peaks appears on the high detuning region (lower detection energies). Although broad (≈ 35 meV), these peaks are nevertheless much thinner than the NRPL spectrum. In the presence of an applied field, each of the roughly equidistant broad peaks that appears on the high detuning region splits into two components, the maximum of which vary fairly linearly with increasing B and are located symmetrically with respect to the energy position of the zero field maximum. Note that the field-ascending and field-descending components associated to neighbour zero field peaks overlap at B ≈ 28T.

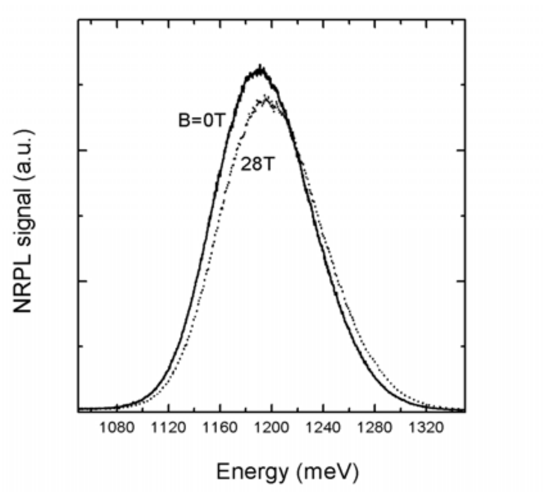

Fig. 1: PL spectrum under non-resonant excitation in the GaAs barrier at zero and strong magnetic field.

3. Discussion and conclusion

We present in what follows an interpretation of our results, which is based on the strong influence of dopants on the hole dot levels. Let us consider initially the large width of the NRPL spectrum. We interpret the NRPL profile in terms of three different contributions: (i) a relatively thinner (≈ 35 meV, peaked around 1190 meV) central part, which is due to the ground S_h-S_e transitions of the dot ensemble and whose width reflects both the usual inhomogeneity observed for undoped dots grown under similar conditions plus a smaller contribution coming from the effect of the dopants on the S_h levels; (ii) a high energy side, related to high-energy transitions which would be forbidden for an isolated QD but that become allowed for the dots perturbed by the ionised impurities ; (iii) a low energy side that results from the superposition of various phonon replica (which become enhanced by the dopants proximity) of the central and high energy parts. Thus, according to this interpretation, the *intrinsic* broadening of the NRPL spectrum of our doped sample is only slightly affected by the doping itself, but the larger observed broadening results from perturbations on the hole levels by the ionised centres nearby the dots.

In order to corroborate our interpretation, we consider in the following the influence of the dopants-related perturbations on the bound dot levels. In fact, the excited (P_h) and ground (S_h) hole states should be rather sensitive (possibly much more than the P_e and S_e electron states) to the perturbations introduced by the ionised dopants nearby the dots. This perturbation leads to various effects on the optical spectra. First, an additional broadening of the inter-band lines is expected, because of the spread of the hole levels. We estimate this inhomogeneous broadening to be at most ≈ 10 meV, thus much smaller than the usual one due to the dots size dispersion. Another consequence is that the P_h-Se and S_h-P_e transitions become optically authorised, in addition to the S_h-S_e and P_h-P_e ones. This is because the dopants induce a mixing of the S_h and P_h states. A PL signal from an excited P_h-S_e state can then arise if the rate for energy relaxation ($P_h \to S_h$) is smaller than the one for radiative recombination. Such transitions lead to a contribution to the NRPL that is, of course, placed on the high energy side, with respect to the ground S_h-S_e one. Finally, the perturbed electron/hole states that contribute to the luminescence have larger couplings to phonons. In fact, a measure of the coupling between confined carriers and optical phonons is usually discussed in terms of the so-called Huang-Rhys factor S. This coupling leads in particular to the appearance of replica in the low-energy side of the photoluminescence signal. However, S is generally small for the ground level of a non-perturbed QDs. Nevertheless, as is well known [5], S increases with increasing dissymetry between the electron and hole probability distributions, and thus becomes sensitive to any perturbation affecting differently the ground electron and hole levels. Various replica are clearly seem in the RPL spectrum in figure 2, which is associated to a class of dots selected by the resonant excitation. The low energy tail of the NRPL spectrum contains the sum of the replica of the whole ensemble of dots, which add to form an average, structureless profile.

We assign the low energy features of the RPL spectrum to phonon replica from the ground dot states. This interpretation allows us to explain also their field evolution in figure 2, as follows. In fact, we assume that the excitation at 1292 meV creates essentially P_h-P_e pairs, which relax down to the ground level S_h-S_e. The dots which match the resonant condition for excitation at B = 0 become detuned in the presence of the field because of the orbital Zeeman effect on the P levels. However, excitations of slightly smaller and slightly larger dots (with slightly higher and slightly lower zero-field P transitions respectively) become allowed. In the same way, the S_h-S_e levels of the new classes of dots are also slightly higher or lower than the ones that emit at zero field. This leads to a peak emission that splits with increasing field. Finally, each of the split S_h-S_e transitions contributes with low energy replica that follow rigidly its field evolution.

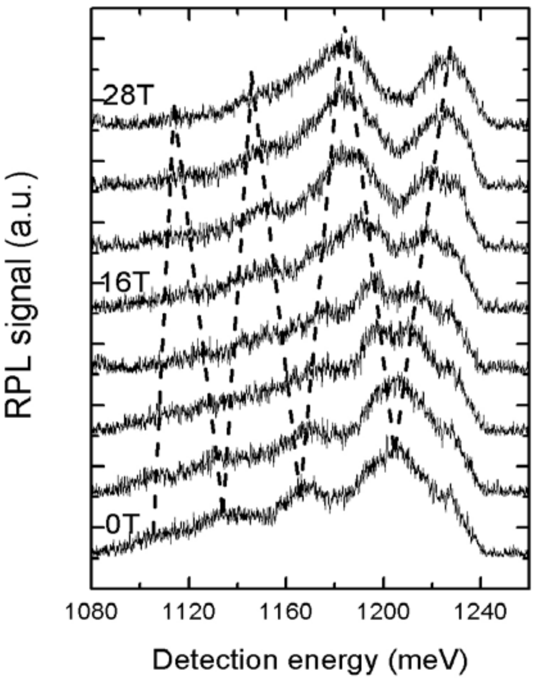

Fig. 2: Magnetic field dependence of the PL spectrum after excitation below the WL edge (at $\hbar\omega_{exc}$=1292 meV). B increases from zero (lower spectrum) up to 28 T (upper spectrum) by ΔB = 4T. The dashed lines are guide for the eyes. The spectra were up-shifted for clarity.

Let us now exclude two others interpretations (currently used for undoped dots) for the results in figure 2, which associate the peaks in the RPL spectrum with electron/hole pairs that recombine in *large* dots selected out of a very inhomogeneous dot ensemble by the resonant laser excitation. In the first interpretation, the energy relaxation involves an integer number of phonons. In this case, the resonant emission of monochromatic LO phonons selects the dots with ground transition detunned from the excitation energy by a multiple of the LO phonon energy (the lower the emitting energy, the larger the average size of the selected dots and the greater the number of phonons emitted). This means that the energy position of the peaks in the RPL spectrum (obtained at fixed excitation energy) would be magnetic field-independent. This trend is in complete disagreement with the results of figure 2, which shows that the energy positions of the low-energy features vary with B, as expected for P states. In the second interpretation, the energy relaxation is not strictly restricted to the emission of an integer number of emitted phonons, and each low energy feature is due to the luminescence of a different class of dots, which have one high energy state at the correct energy for excitation (not necessarily detunned by an integer number of optical phonons from their ground transition). In this case, larger dots contribute to the RPL signal at lower energies. However, since the excitation energy is kept constant, the photoexcitation of large dots would involve high excited transitions (from the D, ... shells), which would then present a larger B-dependent Zeeman shift, as compared to the one for the P states. This trends is also in complete disagreement with the results of figure 2, which shows that all the low-energy features split together with increasing B. Note finally that the replica are less marked at large fields. This is probably due to the fact that the dopant perturbations become less important than the Zeeman corrections for the P states at high fields.

Finally, the weak RPL intensity at small detuning ($\Delta E \leq 52$ meV) is consistent with a fast $P_e \rightarrow S_e$ electron energy relaxation within a dot ensemble with small dispersion for the intra-bande transition $E(P_e)$ - $E(S_e)$. This is corroborated by intraband far infrared absorption measurements performed on the same sample, which display a resonance at about 52 meV, corresponding to the S_e - P_e transition [6].

In conclusion, we have performed in this work NRPL and RPL measurements under strong magnetic field on a sample with charged (n-doped) quantum dots. The analysis of the data provides strong evidences that the broadening of the NRPL line does not give a measure of the QDs size dispersion, and that the coupling of electron/hole pairs with LO phonons is greatly enhanced, because of the presence of the ionised impurities nearby the charged dots.

4. Acknowledgements

The authors thank Drs. G. Bastard and S. Hameau for valuable discussions. The Laboratoire de Physique de la Matiere Condensee de l'Ecole Normale Superieure is "unité associée au CNRS (UMR 8551) et aux Universités Paris 6 et Paris 7".

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